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(54) Title: PROCESS FOR PRODUCING ARTICLES HAVING A SOFT TOUCH

(57) Abstract

The invention relates to a process for producing a product that is pleasant to touch, essentially comprising the single–step injection moulding of a thermoplastic elastomer composition, chosen from the group comprising polyester block copolymers, polyurethanes and styrene block copolymers, which contains glass fibre. Preferably a polyether ester with a Shore D hardness of between 30 and 50 and an MFI of 10–50 g/10 min. containing 35–55 wt.% glass fibre is used in the composition. The product obtained preferably has a stiffness of at least 2000 MPa and a Shore D hardness of less than 60.

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PROCESS FOR PRODUCING ARTICLES HAVING A SOFT TOUCH

The invention relates to a process for producing products having a soft touch. Consumers are imposing increasingly higher demands on articles to be bought by them. In addition to durability and ease of use, an aesthetic appearance, for example, is demanded. Besides design, the latter demand can be translated into a perfect product surface. These aesthetic demands have now evolved to such an extent that, in addition to the eye, the tactile sense has to be pleasantly stimulated when the aforementioned product is handled. Products that must meet the aforementioned demands are for example housings of electric and electronic equipment, for example mobile telephones, remote controls of televisions and other audio and/or visual playing and recording equipment and so-called personal organizers, handles, arms of chairs, steering wheels, switches and keys. These products also demand a good stiffness and must be able to withstand rough treatment.

To obtain products that meet this set of requirements, producers according to the state of the art have to resort to complex production methods that raise costs substantially. For a recent survey of the market and technology reference is made to 'Plastics News' of 21 July 1997, 'Soft touches for soft-touch products'.

A known process is that according to which the product is obtained from a stiff polymer composition by means of injection-moulding and is subsequently coated with a soft polymer that is pleasant to touch. Technical problems that have to be solved are then for example that the polymer composition employed must have both stiffness and ductility and the coating to be applied must show good adhesion to the stiff polymer composition.

This combination of requirements limits the choice of materials. For mobile telephones and the like use is usually made of a stiff polymer composition of acrylonitrile-butadiene-styrene copolymer with polycarbonate (ABS + PC) and a coating of a polyurethane with long alcohol segments.

The use of cheaper (less stiff) polymers, for example reinforced with glass fibre, involves the drawback that they usually show an imperfect surface, as a result of which a thicker coating of the relatively expensive polyurethane is required to conceal these imperfections. The coating is applied with the aid of conventional means or by means of sequential moulding.

Another (known) process is that according to which the surface of the product obtained from a stiff polymer is provided with a fine texture that is pleasant to touch. An example is a texture corresponding to that of an orange peel. This texture can sometimes be applied afterwards, or has to be applied already in a negative form in the wall of the mould in which the product is produced, which is a difficult and cost-raising

operation whose result is generally not considered optimum by consumers.

The inventors have now succeeded in finding a simple process according to which a product having the combination of desired properties indicated above is obtained in a single operation.

The process according to the invention for producing products that are pleasant to touch comprises the single-step injection-moulding of a glassfibre-reinforced thermoplastic elastomer chosen from the group comprising polyester block copolymers, polyurethane and styrene block copolymers.

The thermoplastic elastomer is preferably a polyether-ester block copolymer. The composition's glass fibre content may vary within a wide range, for example between 20 and 70 wt.%, relative to the overall composition. Preferably the glass fibre content is between 30 and 60 wt.%. Most preferable is a glass fibre content of between 35 and 55 wt.%.

The glass fibre content to be chosen will depend primarily on the stiffness of the product to be realized. Preferably the stiffness is characterized by a flexural modulus of at least approx. 2000 MPa.

Elastomer compositions having such a high glass fibre content are known, but are not marketed for injection-moulding applications.

The type of glass fibre may vary in kind of glass, sizing and dimensions. The type chosen will depend primarily on the mechanical properties to be realized. To this end the fibre is usually provided with

a sizing that ensures a strong bond between the glass fibre and the thermoplastic elastomer matrix. A person skilled in the art will easily be able to make this choice from the glass fibres available on the market on the basis of specific experiments.

The aspect ratio (length/diameter) of the fibre in the composition is preferably chosen to be as high as possible, for example at least about 20; preferably the aspect ratio is at least 30, more preferably at least 40. These preferred aspect ratios do however not preclude the possibility of obtaining satisfying results for specific applications with glass fibres having a lower aspect ratio, too. The fibres' diameter is usually between 1.5 and 35 μm, fibres having a diameter of < 15 μm being preferred.

In producing for example housings of electric and electronic equipment it is in many cases advantageous for the composition to contain also fibres of a material that absorbs electromagnetic radiation, for example steel fibres. These fibres are added in an amount such that sufficient protection is obtained. This is determined separately in each individual case.

The composition may also contain the usual additives, for example stabilizers against thermal oxidative influences and against the influence of light, processing auxiliaries, for example flow-promoting agents, in particular polytetrafluoroethylene, colourants and pigments, fillers, for example clay and/or talc and optionally flame retardants.

The thermoplastic elastomer used as the

matrix material is chosen from the group comprising polyurethanes, styrene block copolymers and polyester block copolymers. These polymers are described for example in Chapters 2, 3 and 4 and 8 and 9, respectively, of the book 'Thermoplastic Elastomers', 2nd Ed., Hanser Verlag, Munich (1996) ISBN 1-56990-205-4, to which reference is made for detailed information.

For practical reasons, in particular the highly critical processing conditions, for example the requirement of a low moisture content and the long time that is required after the processing for the product to obtain approximately its ultimate mechanical properties, thermoplastic polyurethane elastomers are however not preferable.

While they are being processed, styrene block copolymers present the problem that the melt viscosity is highly dependent on the shear force and is very high especially at low shear forces, as a result of which processing via injection moulding, though possible, imposes very high demands.

Therefore polyester block copolymers are preferred, more in particular the polyether-ester block copolymers. They may optionally be mixed with a crystalline polyalkylene ester, for example polybutylene terephthalate (PBT). It has most surprisingly been found that a product that is pleasant to touch and has a perfect surface is obtained with these thermoplastic elastomer compositions in one single-step injection-moulding operation. The stiffness of the product obtained in injection-moulding can be varied by

modifying the elastomer matrix and the glass fibre content.

The choice of the polyether-ester block copolymer presents the additional advantage that a relatively large degree of variation in stiffness can be realized without the surface properties varying substantially. On account of the crystallisation rate, polyether-ester block copolymers comprising hard segments composed of butylene terephthalate or butylene naphthalate units are preferred. As the units for the soft segments, use is preferably made of alkylene oxide units having 2 to 8 C atoms; preferably the soft segments are composed of predominantly propylene oxide and/or butylene oxide units. Block copolymers based on ethylene oxide present the drawback that they can absorb large amounts of water and the product obtained therefrom hence shows inferior dimensional stability. For the same reason the polyether-ester block copolymers are preferable to the polyester-ester and the polyether-amide block copolymers.

Preferably the molecular weight of the soft segments is chosen to be between 400 and 3000 and the soft segment concentration between 10 and 65 wt.%, preferably between 20 and 60 wt.% of the polyether-ester block copolymer. The Shore D hardness of the matrix may then vary between 30 and 75, preferably between 30 and 50 (measured according to ISO R 868). A very great advantage in this context is the unusual phenomenon that, in spite of a high glass fibre content, the hardness of the reinforced softer polyether esters is

only slightly higher than that of the non-reinforced, and the high stiffness required can be realized without appreciable concessions with respect to the pleasant tactile properties.

The mol. weight of the thermoplastic elastomer is preferably chosen so that the melt viscosity, expressed as melt flow index, MFI, measured at 230°C and 2.16 kg according to ASTM D1238, in g/10 min., is between 50 and 10, preferably between 30 and 15; most preferable is an MFI of about 25 g/10 minutes.

The injection moulding is carried out under the conditions usually applied for the thermoplastic elastomer concerned. These conditions are specified in the product brochures by the raw material supplier.

The glassfibres-containing thermoplastic elastomer composition can be obtained from the composite components in the usual manners, for example by mixing the polymer in the solid phase in the form of a powder or granulate with the glass fibres. This composition can then be fed directly to the injection-moulding machine's extruder. In another process, which is generally preferred, glass fibres and thermoplastic elastomer and optionally other additives are mixed in the melt. The usual melt-mixing equipment can be used for this purpose. Preferably an extruder is used. Glass fibres and thermoplastic elastomer can then be fed to the throat simultaneously; in another embodiment the elastomer is first melted, after which the glass fibres are fed to the melt. In the latter case a greater fibre length can be realized in the composition, which has a

positive effect on the stiffness of the injection-moulded product. Good dispersion of the glass fibres in the product according to the invention is obtained especially when the glass fibres have been mixed into the melt before the injection-moulding already.

The term 'pleasant to touch' is a subjective measure, which can still not be characterized in a single physical parameter, in spite of extensive research. Parameters such as roughness, hardness and the surface's resistance to friction play a part in this respect. Producers of consumer articles therefore use so-called touch panels to obtain the most accurate possible average evaluation of a surface. Such a qualitative evaluation will be used in the examples in this application.

Experimental

Unless otherwise indicated, the compositions were obtained by mixing with the aid of a Werner and Pfleiderer ZSK 53 twin-screw extruder. The glass fibre was fed to the already melted polymer. The compositions obtained were injection-moulded to form specimens for mechanical tests, and mobile telephone housings for tests by a panel of users. Mobile telephone housings obtained according to the state of the art, i.e. by means of sequential moulding of ABS/PC and a coating of polyurethane and by means of injection moulding of ABS/PC with a textured surface, were also tested by the panel.

Materials employed

Hytrel® polyether-ester block copolymer[1)], Shore D hardness 52, from DuPont de Nemours

Arnitel®E, polyether-ester block copolymer[1)], Shore D hardness 40-55, from DSM

Arnitel®P, polyether-ester block copolymer[2)], Shore D hardness 38, from DSM

Elastollan® 1185A10 polyether-urethane block copolymer, Shore D hardness 39, from Elastogram/BASF (MFIs 90 and 8, resp.)

Glass fibre: OCF®429YZ from Owens-Corning, l = 4.5 mm; d = 10 μm.

The invention will now be further elucidated with reference to the following examples and comparative examples, without however being limited thereto.

Examples

Test methods.

The surface of the products obtained was characterized with the aid of the following tests:

Hardness: Shore D according to ISO R 868

Perfection: by visual inspection

+ no appreciable observable irregularities

0 a few imperfections

- irregular surface

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1 based on polybutylene terephthalate hard segments and tetramethylene oxid soft segments.

2 based on polybutylene terephthalate hard segments and thylene-oxide-terminated polypropylene oxide soft segments

Tactile properties: this subjective evaluation was objectified as much as possible by averaging the evaluations of a panel of users.

+ pleasant according to at least 80% of the panel members

0 indifferent

- has negative aspects according to at least 80% of the panel members

Flexural properties: flexural modulus ISO 178

Impact resistance: Izod notched ISO R 180 (23°C)

MFI: ASTM D1238.

The tested compositions and results are presented in Table I.

Comparative experiments

Telephone housings obtained according to the state of the art, i.e.

A) ABS + PC coated with a polyurethane with long alcohol segments and

B) ABS + PC with a textured surface, were evaluated as (+) and indifferent (0), respectively, by the panel members.

It was commented that the telephone housing with a textured surface tended to slip from the hands.

Table I

Polymer	Glass fibre content [wt.%]	MFI [gr/10 min.]	Shore D hardness	Flexural modulus [Mpa]	Impact resistance [KJ/m²]q	Surface		Comments
						perfection	touch	
Hytrel 5557M (D55)	20	43* (230°C)	64	1190	20	0	+	insufficiently stiff
	30		70	2240	18	0	0	
Arnitel EL550 (D53)	34	25*	63	3000	23	+	+	
EM400 (D38)	19	26*,18 (230°C)	44	780	30	+	+	insuficiently stiff
	34	16	45	1320	26	+	+	
	46,6	13	48	2410	24	+	+	
EM460 (D45)	31,4		52	1920	22	+	+	
	37,4		54	2236	21	+	+	
EL740 (D70)	0	17* (240°C)	70	1045	12	+	+	insufficiently stiff

Polymer	Glass fibre content [wt.%]	MFI [gr/10 min.]	Shore D hardness	Flexural modulus [Mpa]	Impact resistance [KJ/m²]q	Surface		Comments
						perfection	touch	
PL380 (D38)	30	20*,15 (230)	46	1487	27	+	+	
	30	13	52	2121	25	0	+	
	50	10	58	2800	23	0	+	
Elastollan (D39)	30	90* (190)**	50			+	+	
	30	8* (190)	49			-	0	

* MFI non-reinforced polymer according to supplier's pecifications

** The unmarked values measured in the composition.

CLAIMS

1. Process for producing a product having a soft touch, comprising the single-step injection moulding of a thermoplastic elastomer composition, chosen from the group comprising polyester block copolymers, polyurethanes and styrene block copolymers, which contains glass fibre.

2. Process according to Claim 1, characterized in that the thermoplastic elastomer is a polyether-ester block copolymer.

3. Process according to Claim 1 or Claim 2, characterized in that the glass fibre content is 20-70 wt.%, relative to the overall composition.

4. Process according to Claim 3, characterized in that the glass fibre content is 35-55 wt.%, relative to the overall composition.

5. Process according to any one of the above claims, characterized in that the hardness of the thermoplastic elastomer expressed as Shore D lies between 30 and 75, preferably between 30 and 50.

6. Process according to any one of the above claims, characterized in that the melt viscosity of the thermoplastic elastomer, expressed as melt flow index, MFI, measured at 230°C and 2.16 kg according to ASTM D1238, in g/10 minutes, is between 50 and 10, preferably between 30 and 15.

7. Product obtained by the process according to any one of the above claims, characterized in that its stiffness expressed as the flexural modulus is at

least 2000 MPa.

8. Product obtained by the process according to any one of Claims 1-6, characterized in that its Shore D hardness is less than 60.

INTERNATIONAL SEARCH REPORT

Inter nal Application No
PCT/NL 98/00522

A. CLASSIFICATION OF SUBJECT MATTER
IPC 6 C08L53/00 C08L75/04 C08L87/00 C08K7/14

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC 6 C08K C08L

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	DATABASE WPI Section Ch, Week 7522 Derwent Publications Ltd., London, GB; Class A23, AN 75-36698W XP002064705 & JP 50 008850 A (TOYO SPINNING CO LTD) , 29 January 1975 see abstract	1-5
X	PATENT ABSTRACTS OF JAPAN vol. 13, no. 299 (M-847), 11 July 1989 & JP 01 090714 A (INOUE MTP CO LTD), 7 April 1989 see abstract	1,3,4
X	DE 23 50 852 A (BASF AG) 17 April 1975 see page 6; examples 1-5	1,3,4

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[X] Further documents are listed in the continuation of box C.

[X] Patent family members are listed in annex.

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Date of the actual completion of the international search	Date of mailing of the international search report
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C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication,where appropriate, of the relevant passages	Relevant to claim No.
X	DE 37 39 122 A (PEBRA GMBH) 1 June 1989 see column 1, line 63 - line 67	1
X	DE 39 38 891 A (PHOENIX AG) 31 May 1990 see claims 1,6	1,3
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INTERNATIONAL SEARCH REPORT

...formation on patent family members

Inter nal Application No
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